

## Influence of the laser parameters on the space and time characteristics of an aluminum laser-induced plasma<sup>☆</sup>

O. Barthélemy<sup>a</sup>, J. Margot<sup>a,\*</sup>, M. Chaker<sup>b</sup>, M. Sabsabi<sup>c</sup>, F. Vidal<sup>b</sup>, T.W. Johnston<sup>b</sup>,  
S. Laville<sup>b,1</sup>, B. Le Drogoff<sup>b,1</sup>

<sup>a</sup>Université de Montréal, Département de physique, C.P. 6128, Succ. Centre-ville, Montréal, Canada QC H3C 3J7

<sup>b</sup>Institut National de la Recherche Scientifique-Énergie, Matériaux et Télécommunications, 1650 boul. Lionel Boulet, Varennes, Canada QC J3X 1S2

<sup>c</sup>National Research Council of Canada, Industrial Materials Institute, 75 boul. de Mortagne, Boucherville, Canada QC J4B 6Y4

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### Abstract

In this work, an aluminum laser plasma produced in ambient air at atmospheric pressure by laser pulses at a fluence of 10 J/cm<sup>2</sup> is characterized by time- and space-resolved measurements of electron density and temperature. Varying the laser pulse duration from 6 ns to 80 fs and the laser wavelength from ultraviolet to infrared only slightly influences the plasma properties. The temperature exhibits a slight decrease both at the plasma edge and close to the target surface. The electron density is found to be spatially homogeneous in the ablation plume during the first microsecond. Finally, the plasma expansion is in good agreement with the Sedov's model during the first 500 ns and it becomes subsonic, with respect to the velocity of sound in air, typically 1 μs after the plasma creation. The physical interpretation of the experimental results is also discussed to the light of a one-dimensional fluid model which provides a good qualitative agreement with measurements.

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### 1. Introduction

Laser-induced plasmas at low fluence (typically 10 J/cm<sup>2</sup>) have various applications such as pulsed laser deposition

(PLD) or multi-elemental analysis. The latter technique, known as Laser-Induced Plasma Spectroscopy (LIPS) or Laser-Induced Breakdown Spectroscopy (LIBS), consists in analyzing the light spectrum emitted from a plasma created at the sample surface by means of laser pulses, most of the time in ambient air. LIPS has many practical advantages over more conventional elemental analysis techniques and is consequently being considered for a growing number of applications [1,2] including on-line process or in situ analysis. A more detailed description of this technique and its main characteristics can be found elsewhere [3–5].

Despite the numerous applications of LIPS, actual and potential, the knowledge of laser-induced plasmas in ambient air remains scarce and their modeling extremely limited. For this reason, over the last few years, our group has invested in a deeper understanding of such plasmas by using a combined experimental and theoretical approach [6–14]. Many of these publications deal with the experimental

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\* Corresponding author. Tel.: +1 514 343 6635; fax: +1 514 343 2071.

E-mail addresses: olivier.barthelemy@umontreal.ca (O. Barthélemy), joelle.margot@umontreal.ca (J. Margot), chaker@emt.inrs.ca (M. Chaker), mohamad.sabsabi@cnrc-nrc.gc.ca (M. Sabsabi), vidal@emt.inrs.ca (F. Vidal), johnston@emt.inrs.ca (T.W. Johnston), stephane.laville@cnrc-nrc.gc.ca (S. Laville), boris.ledrogoff@cnrc-nrc.gc.ca (B. Le Drogoff).

<sup>1</sup> Present address: National Research Council of Canada, Industrial Materials Institute, 75 boul. de Mortagne, Boucherville, Canada QC J4B 6Y4.

investigation of the influence of the pulse duration on the plasma characteristics, whereas others are related to a plasma model predicting the dependence of target ablation and plasma properties on laser conditions. Because the assumption of local thermodynamic equilibrium (LTE) condition is essential to characterize the plasma, a specific experimental study has also been devoted to this question [13].

Most published papers related to laser plasma characterization [15–19], including ours [7,10,11,13], provide space-integrated measurements. Only a few studies report observations of the ablation plume with spatial resolution. These publications deal with time-integrated laser-induced plasmas in ambient air [20] as well as in controlled atmospheres (Ar, He, O<sub>2</sub>, N<sub>2</sub>), at either atmospheric pressure [21,22] or low pressure [23–26] (see also the review by Capitelli et al. [27] and references therein). Among the small number of investigations that were conducted in ambient air with both space and time resolution, in one [28–30] or two space dimensions [31–36], spatial profiles of the plasma parameters are only provided at a few delay times after plasma creation, making difficult to achieve a complete picture of the plume evolution.

In the present paper, our goal is to perform space- and time-resolution of the plasma parameters in the most usual laser conditions relevant to LIPS, i.e., at low fluence in ambient air. More specifically, we investigate the dependence of plasma electron density and temperature one-dimensional (1-D) profiles (i.e., axial profiles with integration on the transverse coordinate) on the laser pulse duration (ranging from fs pulses to ns pulses) and wavelength (from ultraviolet (UV) to infrared (IR)), together with their time evolution. From these profiles, we were able to characterize plasma expansion length and velocity.

Section 2 is devoted to the experimental setup and procedure used in this work to achieve space- and time-resolved observation of the plasma. In Section 3, we briefly present the spectroscopy diagnostics employed to measure the electron density and temperature. Experimental results are shown in Section 4 while Section 5 presents simulation results related to the experiments. Finally, Section 6 concludes this article.

## 2. Experimental setup and procedure

The experimental setup is shown in Fig. 1. It is similar to that employed by Monge et al. [31]. The laser beam is focused on a spot of 0.4 mm<sup>2</sup> perpendicularly to an Al target surface, using a quartz lens ( $f=25$  cm). The target is an aluminum alloy standard sample from Alcan containing 0.65% of iron. Note that this iron concentration is sufficiently small (0.65%) to weakly influence Al plasma characteristics.

The plasma created by two types of lasers was investigated. One is generated by a Nd:YAG laser emitting 6 ns pulses at the fundamental wavelength ( $\lambda_L=1064$  nm), as well as the second ( $\lambda_L=532$  nm) and fourth harmonics ( $\lambda_L=266$  nm). This system can therefore be used to study the influence

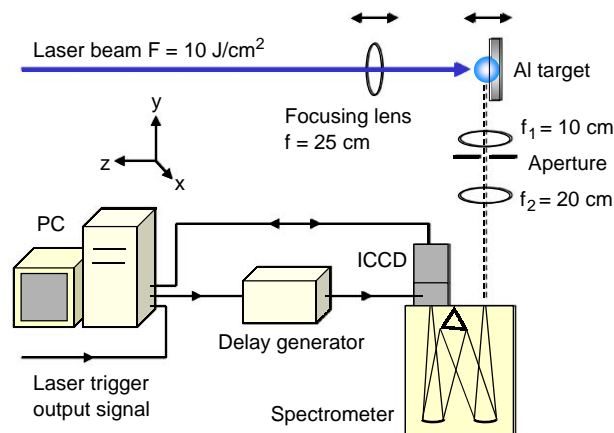


Fig. 1. Experimental setup used for spectroscopic measurements with time and spatial resolution.

of the laser wavelength on the plasma properties. The second laser is a tunable chirped-pulse amplifier Ti:Sapphire laser emitting at  $\lambda_L=800$  nm. As the pulse duration  $\tau$  of this laser can be varied, the influence of this parameter on the plasma characteristics was investigated for  $\tau=80$  fs, 3 ps and 270 ps. For both Nd:YAG and Ti:Sapphire lasers, the focusing parameters were kept the same yielding in every case a focal spot of 0.4 mm<sup>2</sup>, while the pulse energy was set at 40 mJ, yielding a fluence  $F=10$  J/cm<sup>2</sup> (irradiances ranging from 1.7 GW/cm<sup>2</sup> for 6 ns pulses to  $1.3 \times 10^5$  GW/cm<sup>2</sup> for 80 fs pulses). Finally, the repetition rate was held to 2 Hz.

As shown in Fig. 1, the light emitted by the plume is collected by a set of two lenses ( $f_1=10$  cm and  $f_2=20$  cm) along a direction perpendicular to the laser beam axis (i.e., parallel to the target surface) and focused onto the entrance slit of an imaging spectrometer. The collection system employed yields a magnification of 2. An aperture (diameter of 1.5 cm) placed just behind the first lens is used to reduce spherical aberrations that cause image blur, which results in an improvement of the spatial resolution. The spectrometer is a Jobin-Yvon Triax 550 (focal length 55 cm) equipped with a 3600 g/mm grating and an intensified Charged Coupled Device (CCD) camera (Andor Technology). The width of the entrance slit of the spectrometer was set at 50  $\mu$ m. A delay generator triggered with a signal provided by the laser was used to time-shift the CCD gate with respect to the laser pulse, therefore allowing time-resolved measurements.

In Fig. 1, the laser beam axis is identified as the  $z$ -axis and the spectrometer optical axis as the  $y$ -axis. In this configuration, the spectrometer entrance slit collects the light emitted by a “slice” of plasma of thickness  $\Delta z$  along the  $(x,y)$  plane. As a result, the measurements performed with this setup yield values that are space-averaged over the whole volume of the considered plasma slice. One-dimensional resolution of the ablation plume is achieved by imaging various plasma slices onto the entrance slit when moving the target along the  $z$ -axis. In order to keep the same laser beam focusing parameters (spot size and fluence), the

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